SEMI-ANNUAL STATUS REPORT #2

For the Period

1 January 1967 to 30 June 1967

DEFECT PRODUCTION IN SINGLE CRYSTALS RESULTING FROM ION BOMBARDMENT

by

Lawrence B. Shaffer

Prepared for

National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio

31 July 1967

GRANT NGR 36-019-001

X RAY PHYSICS LABORATORY
Hiram College
Hiram, Ohio 44234

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I. INTRODUCTION AND SUMMARY

Contained in this report is a summary of the progress made during the period of time from January through June, 1967 on work supported through the Lewis Research Center of The National Aeronautics and Space Administration. Initiated in June, 1966 this research program includes two tasks: the first is a study of the damage produced on the surface of metal single crystals during ion bombardment using x-ray methods and the second is a determination of certain thermodynamic properties of cesium gas using x-ray methods. A possible further area of research is a study of cesium gas as it is adsorbed onto metal single crystals as a special case of interest in gas-solid interactions.[1]

Since most of the first year has been spent designing and assembling the equipment this report will include a discussion of the equipment assembled to date.

II. SINGLE CRYSTAL DAMAGE STUDIES

The final design on the special vacuum chambers was approved and the system has been under construction by Varian Associates since February, 1967. Shipment has been promised April 21, May 5, May 26, June 30, July 14, July 21, and July 28, 1967. The system is now almost four months late and this delay has seriously impeded progress on the research grant. The final system configuration will be reported in detail in the next report.

Also during this past period the requirements for the double crystal x-ray spectrometer were finalized. Since the required design was similar to a spectrometer already available at the Ohio State University Physics Department and since the anticipated use of the spectrometer at Ohio State over the next few years was low, a request was made to Professor Jossem, chairman of the Physics Department, to loan the spectrometer to Hiram College with the understanding that we could make certain modifications to the spectrometer for our use and also that the possibility of some collateral studies should be pursued. This request was approved, the spectrometer is now at Hiram, and most of the modifications have been completed.

III. CESIUM GAS SCATTERING STUDIES

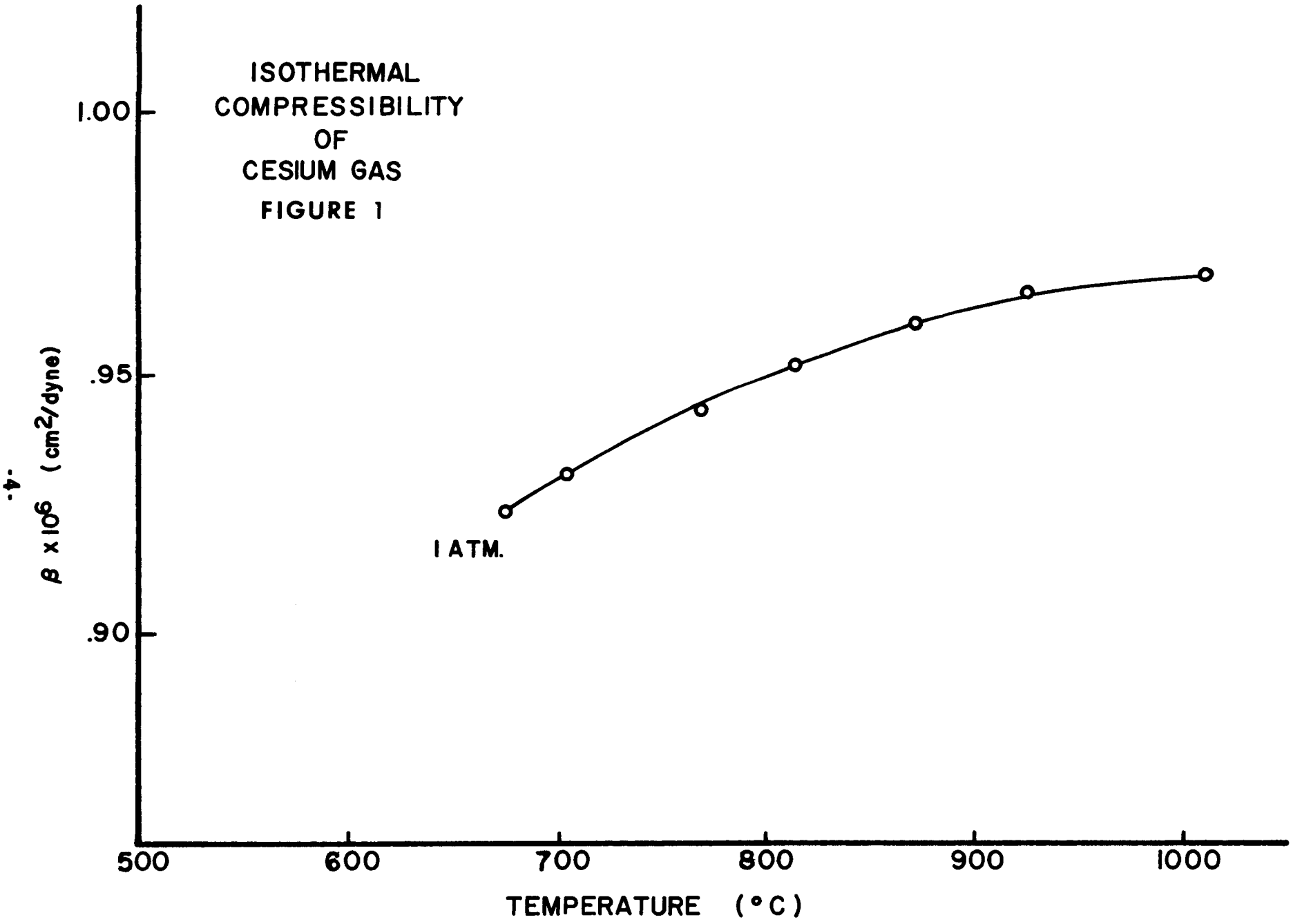
Theoretical and Problem Evaluation Studies

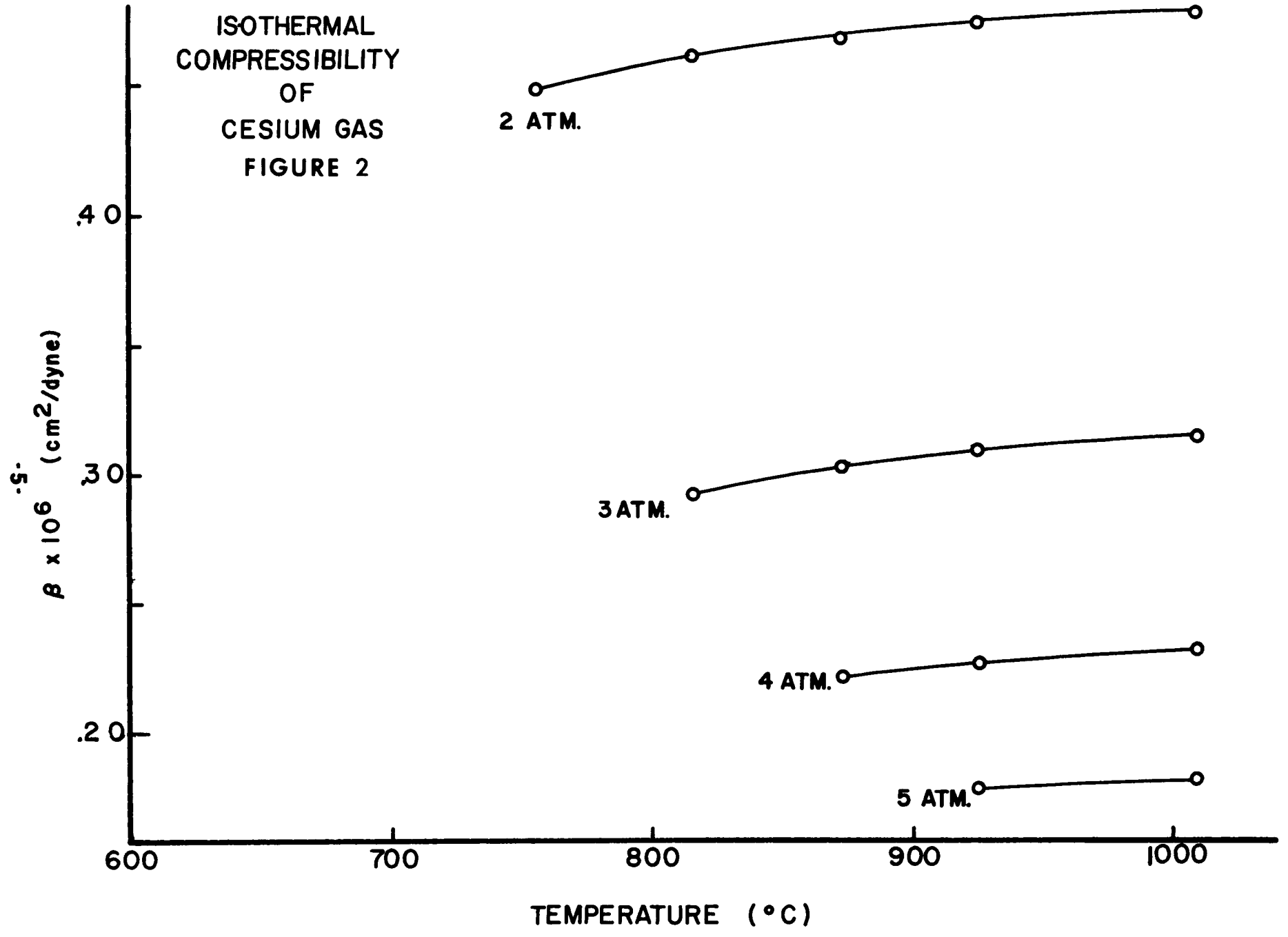
In order to determine the interaction potential of a pair of cesium atoms and to make certain absolute measurements of other thermodynamic properties of cesium vapor, small angle x-ray scattering is to be employed. Calculations on the interaction potential require that the number density c of the vapor be known. This can be determined by measurement of the scattering with x-ray beams of known absolute intensity. The apparatus and procedures to be employed should make such determinations possible. In addition it is planned to make observations on the adsorption of cesium atoms on the walls of containing vessels.

Employing the data of Heimel[2] and Ewing,[3] et al., calculations have been made on the value of the isothermal compressibility and density of cesium vapor in the temperature range from 676°C to 1010°C and in the pressure range from 1 to 5 atmospheres as shown in Figures 1 to 3. This value combined with the number of atoms per cc (number density), the Boltzman Constant k, and the Kelvin Temperature T, yields the product $ckT\beta$, the interparticle interference function at zero scattering angle.[4]

In all calculations in the above range, however, this product is essentially unity which indicates a nearly ideal gas with zero interaction potential. Since it seems reasonably certain from the properties of other vapors that at temperatures near the condensation point such an interaction should become evident, it is planned to make observations at temperatures and pressures very close to the saturation curve of cesium vapor. Since this product contains c, then for a constant c and T the value of $ckT\beta$ should follow β and should the product depart significantly from unity, it would indicate that β does not follow the value predicted for an ideal gas, namely, $\beta = \left(\frac{1}{P}\right)_V \propto \left(\frac{1}{T}\right)_V$.

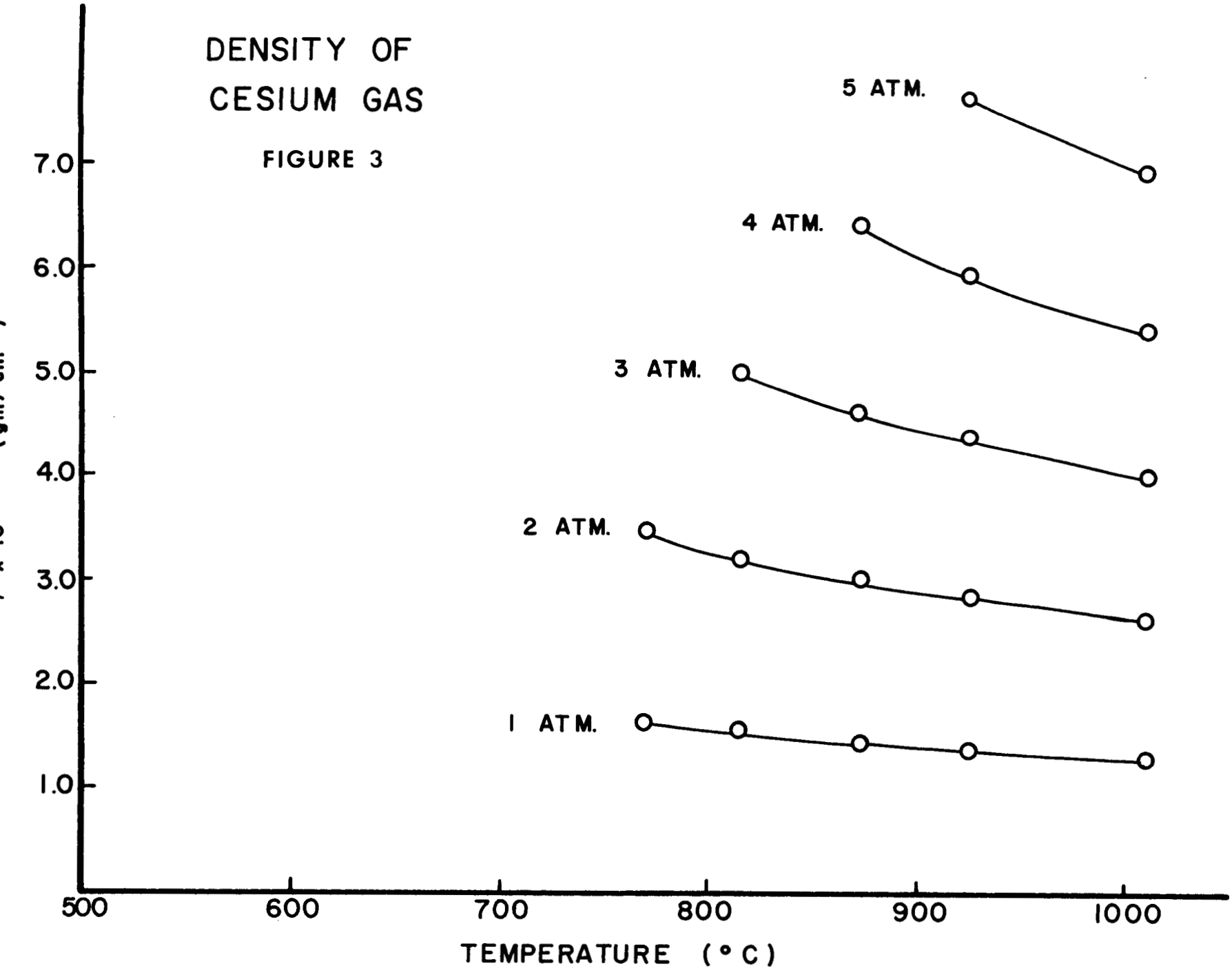
The above consideration assumes a constant c. However if in the process of heating, cooling, and reheating the cesium vapor vessels, the adsorption of cesium atoms on the walls varies to any great extent the value of c would be uncertain. Calculations assuming a continuous monolayer of adsorbed cesium atoms in vessels whose dimensions are comparable to ours indicate that a negligible variation in c could arise with departures from a continuous monolayer (see page 7). Further investigation of adsorption is to be made by


ISOTHERMAL
COMPRESSIBILITY
OF
CESIUM GAS
FIGURE 1
1.00
.95
.90
β x 10^6 (cm^2/dyne)
1 ATM.
500
600
700
800
900
1000
TEMPERATURE (°C)


ISOTHERMAL
COMPRESSIBILITY
OF
CESIUM GAS
FIGURE 2
2 ATM.
3 ATM.
4 ATM.
5 ATM.
.40
.30
.20
600
700
800
900
1000
TEMPERATURE (°C)
β x 10^6 (cm^2/dyne)^-.5

DENSITY OF
CESIUM GAS

FIGURE 3



measuring the resistance across the length of the cesium vapor vessels. Such resistance should be influenced by the completeness of the adsorbed layer and its thickness, assuming more than a monolayer.

Experimental Techniques

Absolute Intensity Measurements

To measure the large ratio ($\sim 10^{-7}$) of the scattered to the incident x-ray beam special techniques must be employed.[5,6,7] The principle investigator gave an invited paper on this topic last January in which three techniques were described.[8] The technique to be used primarily in this study will be that of scattering from a gas sample such as $C_4 F_8$ or SF_6. Then with calculations from the thermodynamic data for three nearly ideal gases, the predicted scattered intensity can be found and compared with the observed scattered intensity to find the absolute intensity calibration constant. However still more study needs to be done on comparing different x-ray geometries and standard samples as well as on the effects of all the necessary corrections to the data. The principle investigator intends to publish more about this technique in the near future and will defer a more detailed discussion until that time.

Sample Holder

Through the courtesy of H. R. Letner of the General Electric Company, we have secured a number of Lucalox (Al_2O_3) tubes with niobium-zirconium alloy end caps such as are being used in a

recently designed lamp. These tubes, I.D. circa 7/32" = 0.555 cm - O.D. circa 16/32", length 3 3/4" plus end caps, are to be filled with saturated cesium vapor at temperatures between 664°C and 1000°C. Our source of information on the Lucalox tubes states that, "all of these materials-Al_2O_3, niobium, and the tungsten coils in the end caps including the sealant used to fasten the end caps to the ceramic-should resist attack by cesium vapor for long periods of time at temperatures of at least up to 700°C to 800°C." Aside from attack by cesium, the structures will withstand several hundred more degrees if not subjected to severe thermal shock.

These tubes will then be sealed off, brought to Hiram College, and reheated to their saturation temperature in an x-ray diffractometer where the small angle scattering they produce will be observed. From the curve of scattered intensity vs. scattering angle it is expected that effects of the atomic interaction potential will be observed and calculations made to evaluate this potential.

Further investigation with these tubes includes an attempt to observe and possibly derive some conclusions as to the adsorption of cesium atoms on the tube walls. This will be done by observation of the resistance of the tubes while heated to various temperatures. Measurements will be made on an empty tube and on those that are filled at the several temperatures.

Although so far the Lucalox tubes seem ideal in many ways, Cs insertion, seal off, high temperature capability, electric discharge capability, etc., two serious problems have been found with their use. First, it has been noted that indium oxidizes completely in a few minutes' exposure to air at, say 800°C.

Hence, throughout this treatment at elevated temperatures, they must be surrounded with an inert atmosphere or vacuum. Second, and more serious, is that the x-ray transmission for CuK_α is very low ($< .005\%$). Thus either the walls must be made thinner, a possibility now under study, or a higher energy x ray must be used such as MoK_α. The transmission of the Lucalox tube for MoK_α is 8%. If the walls can be made thinner, an even higher transmission will result. To obtain MoK_α radiation, a modification to the present x-ray tube must be made involving plating some molybdenum onto the Cu anode.

Cesium Gas Adsorption

The ratio of cesium atoms adsorbed on a monolayer to the number in the vapor state at 676°C and 1 atmosphere in a tube 0.555 cm diameter is calculated as follows:

Ratio of surface area to volume $= \frac{2\pi r}{\pi r^2} \times 1 = \frac{4\pi D}{\pi D_o^2} \times 1 = \frac{4}{D} = 7.23$

Given a monolayer of Cs atoms with a diameter of $5.4\text{Å} = 5.4 \times 10^{-8}$ cm then the area/atom $= \frac{\pi}{4}(5.4 \times 10^{-8})^2 = 23 \times 10^{-16}$ cm^2/ atom and the number of atoms/cm^2 $= \frac{1}{23 \times 10^{-16}} = 4.35 \times 10^{14}$ atoms/cm^2 assuming spherical atoms completely cover the surface. From Heimel[2] the number of atoms/cc at 664°C is 8.23×10^{18} atoms/cc. In a 1 cm length of tube, 0.555 cm diameter, the ratio of the adsorbed atoms in a single monolayer to the number in the 1 cm column of vapor is

$$7.23 \times 4.35 \times 10^{14}/8.23 \times 10^{18} = 3.88 \times 10^{-4} = 0.0388\% = 0.04\%$$

The adsorbed monolayer is therefore a negligible fraction of the vapor and large changes in it would produce no detectable change in the number density.

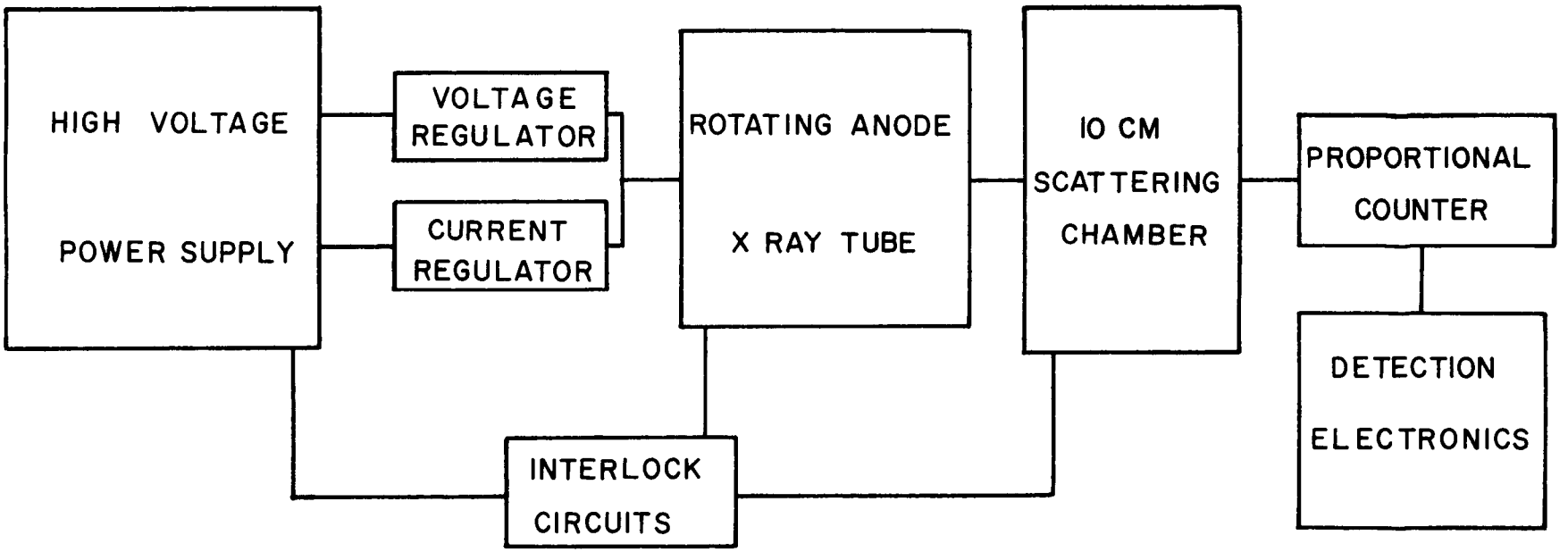
Experimental Facilities

A block diagram of the 10 cm x-ray diffractometer is shown in Figure 4 and each major component will now be discussed in some detail.

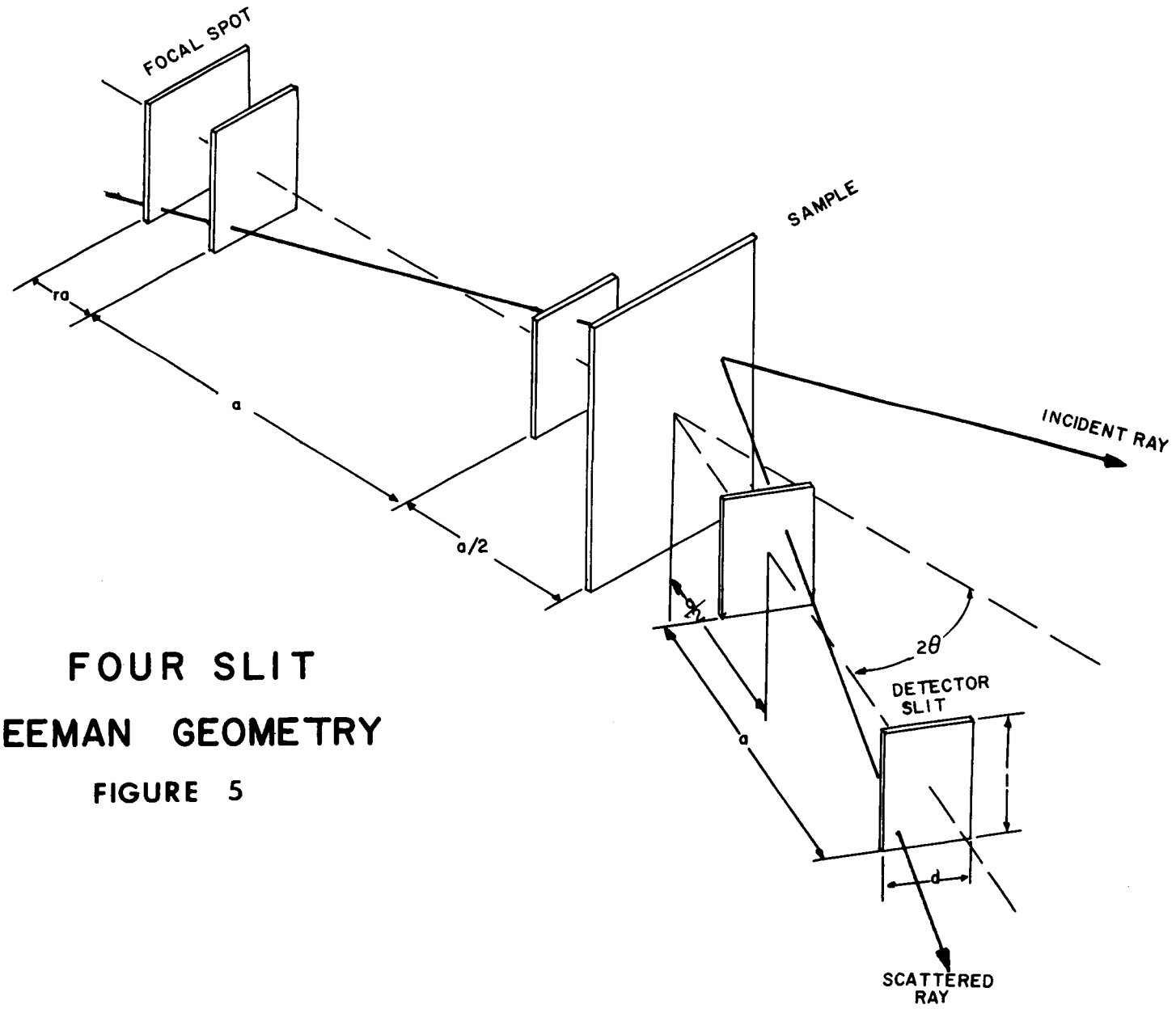
10 cm Scattering Chamber

The definition of the x-ray beam is accomplished by four equally spaced vertical slits.[9] The first two slits are stationary and collimate the x-ray beam incident on the sample which is placed midway between the second and third slits. The last two slits, which are mounted on an arm with the detector tube so as to rotate about a vertical axis through the scattering sample, analyze the scattered x-ray beam as a function of angle as shown in Figure 5. Successive slits are 10 cm apart, have tantalum edges, and are symmetrically adjustable in slit width from 0.050 to 2.000 mm. The slits are all set to the same slit width, usually 0.400 mm, where narrow slit widths give greater angular resolution and a smaller minimum angle (before slit edge scattering is seen) and wider slit widths give greater intensity. The minimum angle at which data can be taken without seeing slit edge scattering is $\epsilon_m = 4\,\frac{d}{a}$ where d is the slit width and a is the slit separation. All slit lengths (vertical direction) are set to 6.0 mm using a surface gauge and vernier calipers. The x-ray focal line is about 17 cm behind the first slit.

The slit system can be completely enclosed in a steel chamber thus allowing for evacuation of the chamber or flooding with helium to reduce both air scattering and air absorption as shown in Figure 6.



BLOCK DIAGRAM OF
10 CM X RAY DIFFRACTOMETER
FIGURE 4



FOUR SLIT
BEEMAN GEOMETRY

FIGURE 5

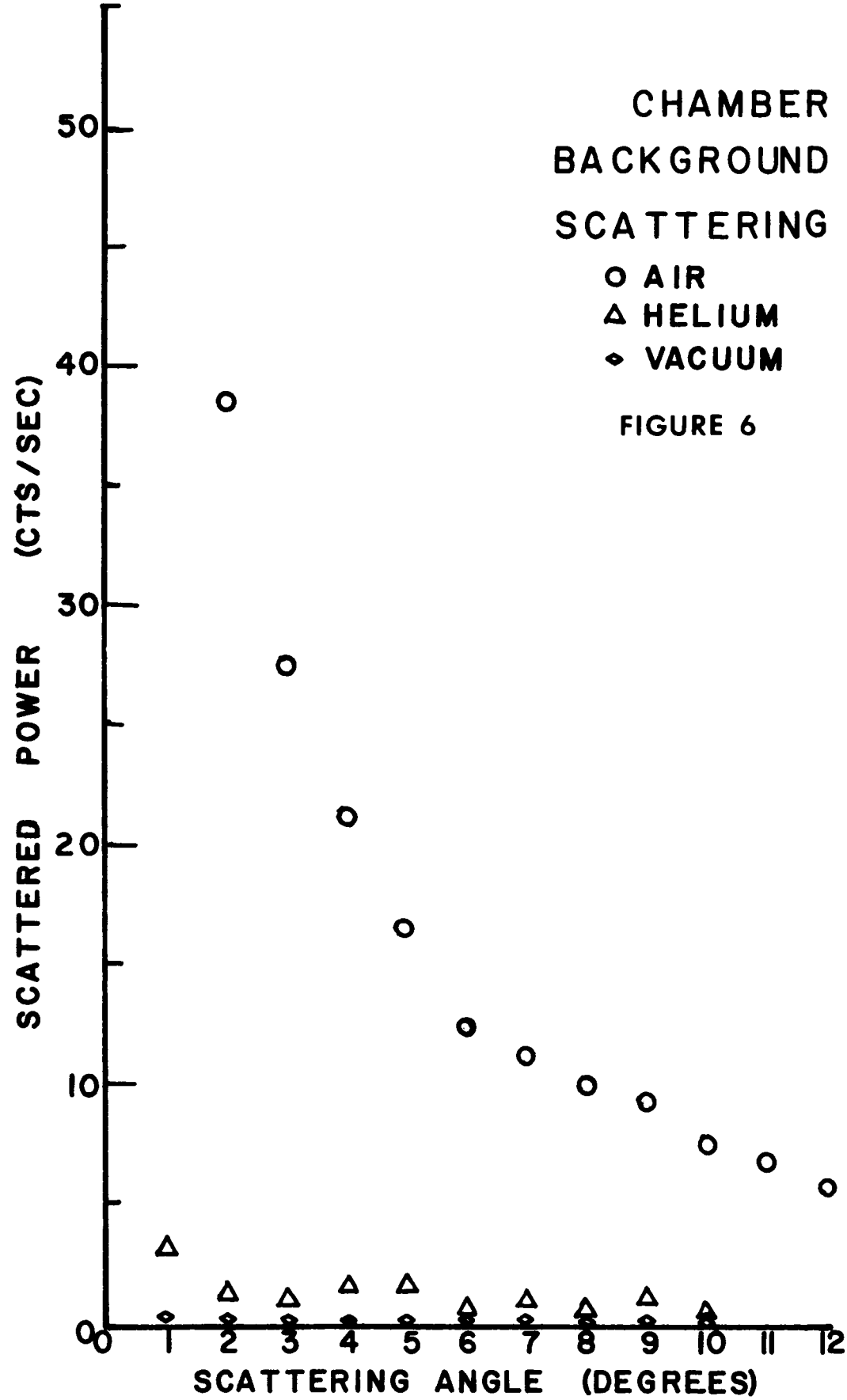

CHAMBER
BACKGROUND
SCATTERING
AIR
HELIUM
VACUUM
SCATTERED POWER (CTS/SEC)
0
10
20
30
40
50
0 1 2 3 4 5 6 7 8 9 10 11 12
SCATTERING ANGLE (DEGREES)

FIGURE 6

With the counter tube set to receive the 9° polystyrene peak, a 21% gain in intensity is observed when He floods the chamber and a 41% gain when the chamber is evacuated. This gain in intensity shows the decrease in the beam absorption as the air in the chamber is flushed out with He or evacuated. The exit window permits analyzing scattering data over the angular range from -37° to 110°. Both the entrance and exit windows are of 1 mil, type S Mylar. The detector tube rides outside the chamber on a supporting arm which is mechanically linked to the arm inside the chamber holding the last two slits.

A hole in the top plate of the steel chamber permits a four position sample holder turret to place the sample in the sample position. This turret, which rotates about a vertical axis, permits changing the sample four times without disturbing the chamber environment. A single position sample holder is also available which mounts in a conical seat midway between the second and third slits.

A special furnace and sample holder are being designed for the Cs gas scattering experiment and the problems so far have been discussed previously. This furnace will be completely inside the steel chamber so that when the furnace is hot the chamber will be evacuated thus reducing thermal insulation problems as well as increasing safety since the Cs will be enclosed inside the 1/2" steel chamber.

X-Ray Detection Electronics

A block diagram of the detection electronics is shown in Figure 7. Except for the high voltage power supply,[10] proportional counter tube,[11]

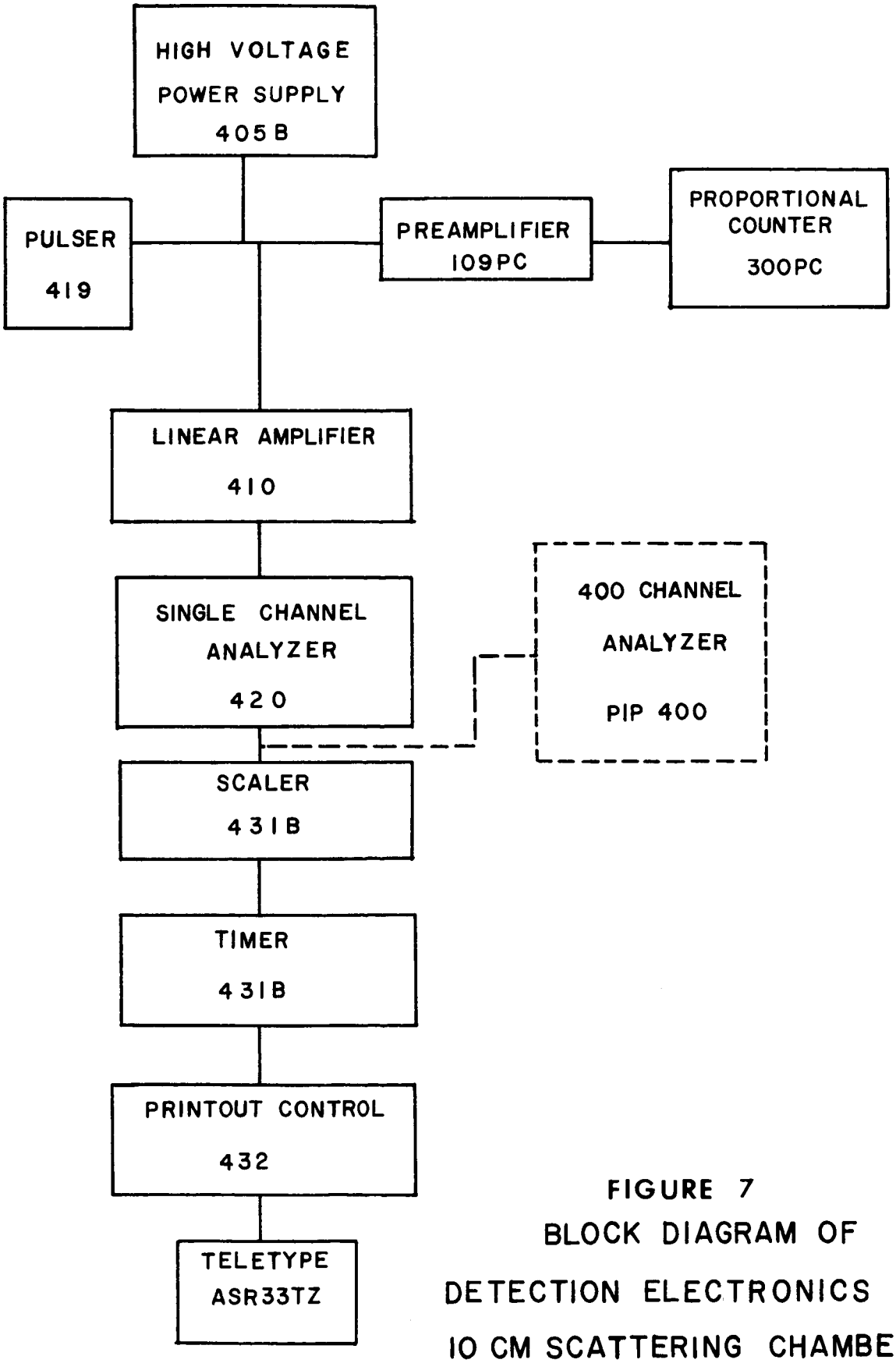


FIGURE 7
BLOCK DIAGRAM OF
DETECTION ELECTRONICS FOR
10 CM SCATTERING CHAMBER

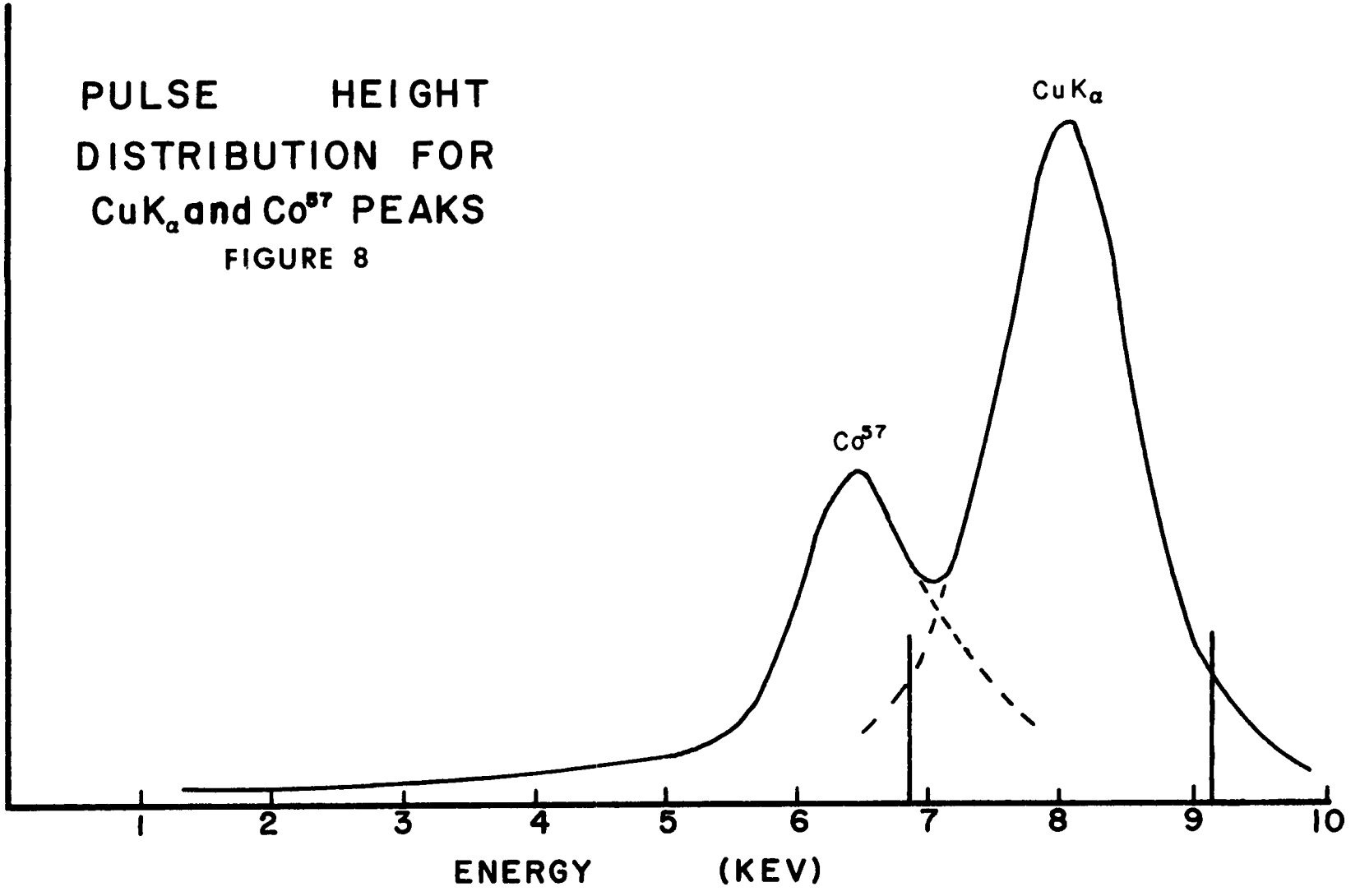
and Teletype[12] all the equipment was purchased from ORTEC[13] and the model numbers for each item are shown on the diagram. All the ORTEC equipment except the preamplifier conform to AEC standards and rack mount in a 401 Bin and 402 Power Supply Unit utilizing only 8 3/4" of rack height.

The proportional counter was chosen for the 10 cm geometry due to its better energy resolution than the scintillation detector[14] (to be used on the double crystal spectrometer). The 410 Multimode Amplifier has gain selection from 0.75 to 1350, single or double differentiated delay line or RC shaping, RC time constants from 0.1 to 10.0 microseconds, simultaneous unipolar and bipolar outputs, etc. The 431B Scaler/Timer module has an internal 0.04% accurate tuning fork standard for operating in the timer mode or functions as a 6 decade 2 MHz scaler in the scaler mode as selected by a front panel switch. The 432 Printout Control will provide the necessary control, format, and sequencing to print out up to 50 Scaler/Timer units into a Teletype Page printer producing both hard copy and punched paper tape for computer input.

This set-up provides a very versatile arrangement for either x-ray spectroscopy or diffraction depending on the use of the amplifier and single channel analyzer or the flexible readout devices. Alignment of the system is easily accomplished using the pulser and 400 channel analyzer.[15] After first calibrating the pulser with 6.4 Kev radiation from Co^{57} and finding the CuK_α peak on the PIP 400, the single channel analyzer can be set using the pulser to simulate the CuK_α peak edges. A typical spectrum showing the CuK_α peak, the 6.4 Kev peak, and the upper and lower discriminator levels is displayed in Figure 8.

PULSE HEIGHT DISTRIBUTION FOR CuK_α and Co^{57} PEAKS

FIGURE 8

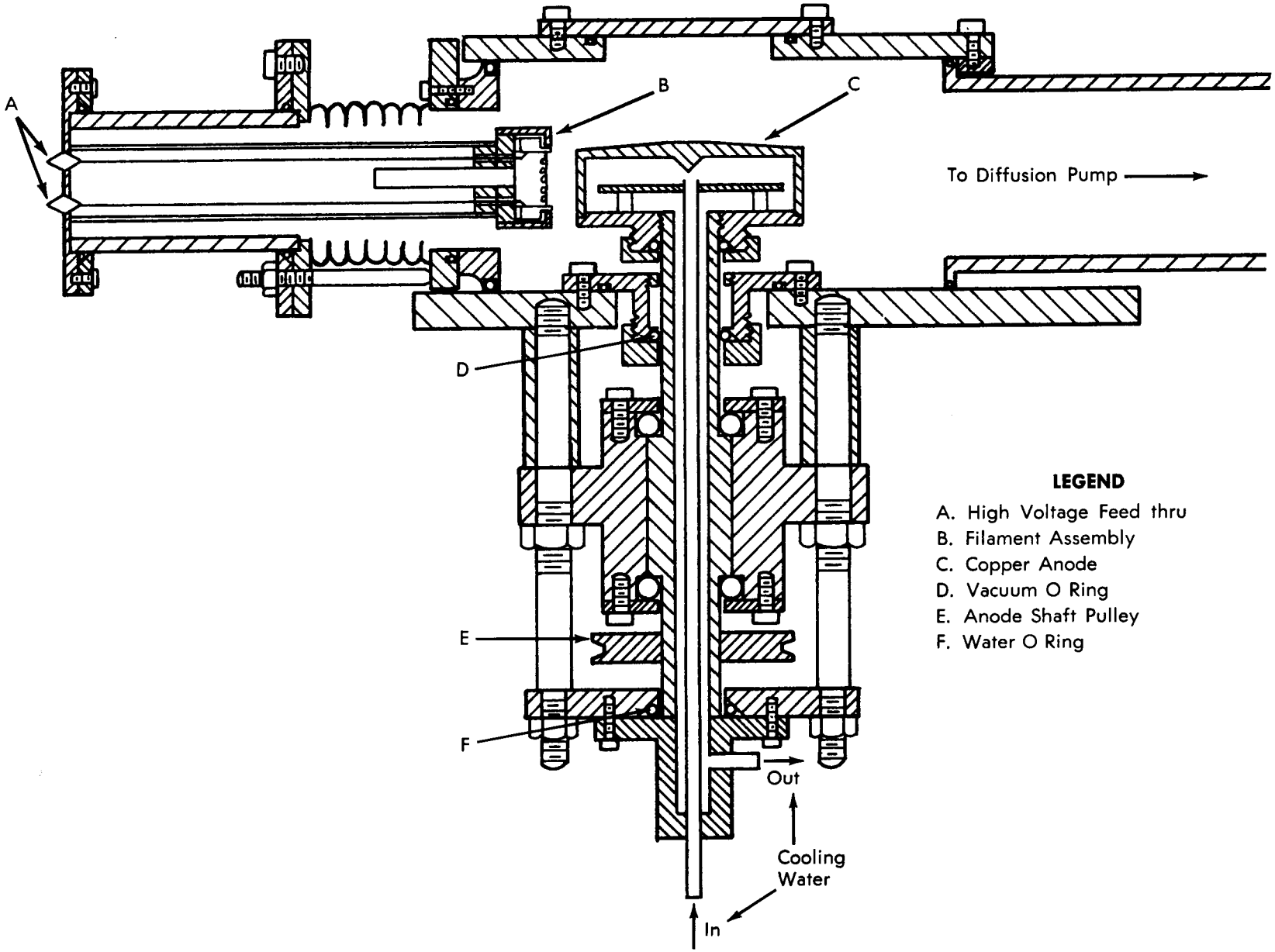


Rotating Anode X-Ray Tube

A high intensity x-ray beam is needed for the 10 cm scattering chamber described above not only due to the long x-ray path, approximately 50 cm, but also due to the weakly scattering samples to be investigated such as Cs gas. Thus a water cooled, continuously pumped, rotating anode x-ray tube has been set up for this purpose. Figure 9 shows a cross sectional view of the x-ray tube the important points being the "O" ring vacuum and water seals for the rotating shaft. Other features of this tube include a demountable filament assembly and demountable anode. Thus repairs on the tube can be made such as replacement of filament, anode, and shaft seal "O" rings. The filament assembly can be mounted in either a horizontal or vertical port giving a vertical or horizontal focal line respectively. In addition the horizontal focal line can be shortened by rotating the filament assembly so that a focal spot is obtained if desired. Anodes of materials other than copper can be used or certain target materials can be plated onto the copper targets (three are available).

Required services for the x-ray anode are:

1. the anode motor to rotate the anode at approximately 920 rpm
2. the water pump and heat exchanges to circulate distilled water to cool the anode
3. the forepump and diffusion pump to maintain a vacuum of less than 2×10^{-5} Torr in the x-ray tube
4. water cooling outside the anode
5. a high voltage power supply.



Rotating Anode X-Ray Tube Schematic

Figure 9

High Voltage Power Supply

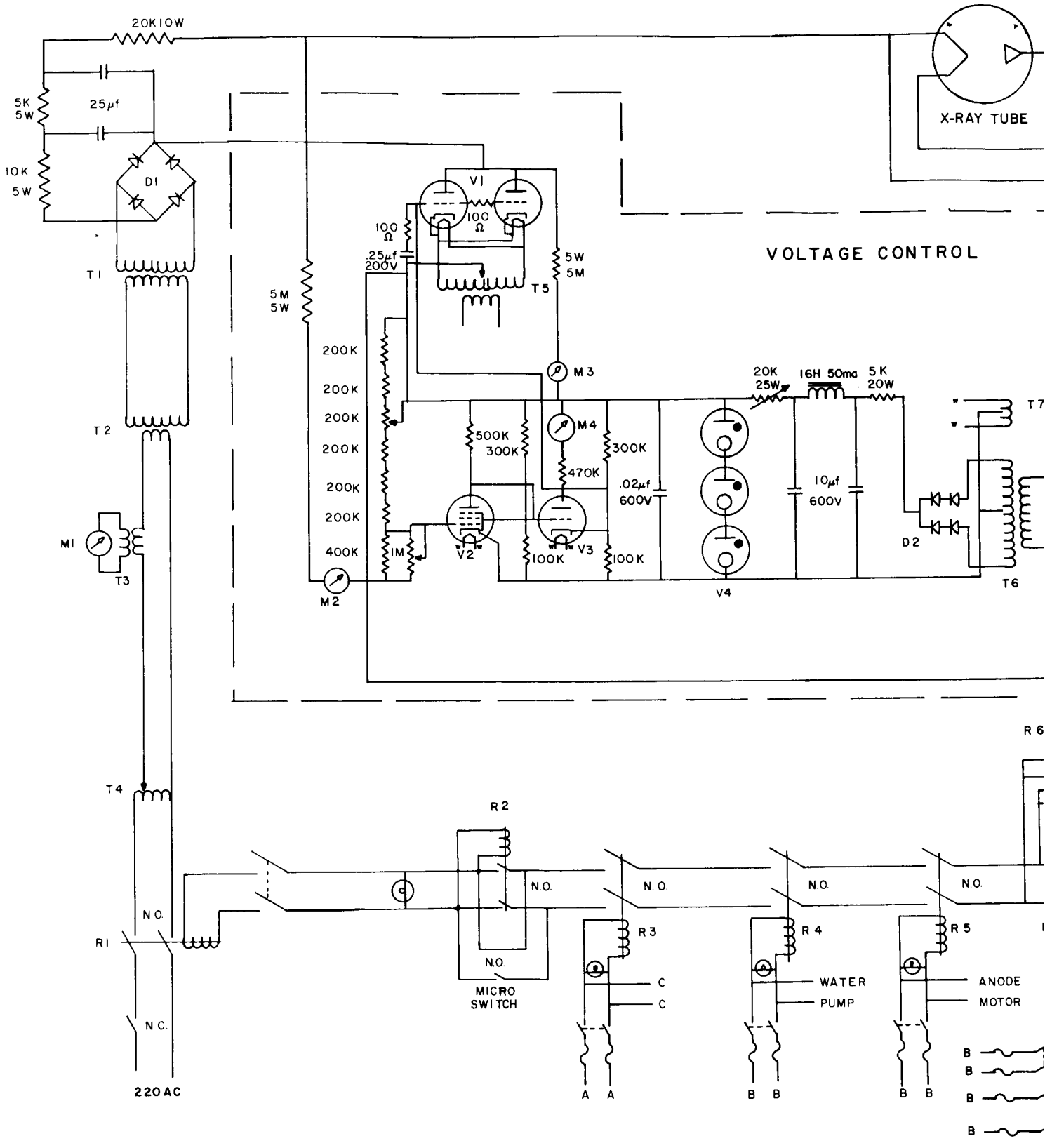
A 50 KV, 200 MA regulated power supply has been constructed to provide the power to the x-ray tube. For the most part, the circuit has not been changed from that previously used[6] and is included here for completeness in Figure 10. Some of the standard features of this power supply are that both current and voltage are continuously variable and that both current and voltage are regulated to $< 0.1\%$ and the unique feature of this power supply is the use of a bridge circuit with four silicon diodes rather than hot filament type diodes.

The power supply also includes vacuum control and interlock circuits as shown in Figure 10. The vacuum control circuit monitors both the high pressure in the x-ray tube and the forepressure and whenever the vacuum goes "bad," automatically shuts down everything to protect the high voltage power supply, x-ray filament, and diffusion pump. The vacuum control circuit also monitors the vacuum in the 10 cm scattering chamber.

The interlock circuits insure that the proper services such as vacuum, cooling water, rotation, etc. are supplied to the anode before the high voltage can be turned on. Also other safety devices are provided such as the high voltage variac which must always be turned up from zero volts.

IV. RESULTS, RECOMMENDATIONS AND PROBLEMS

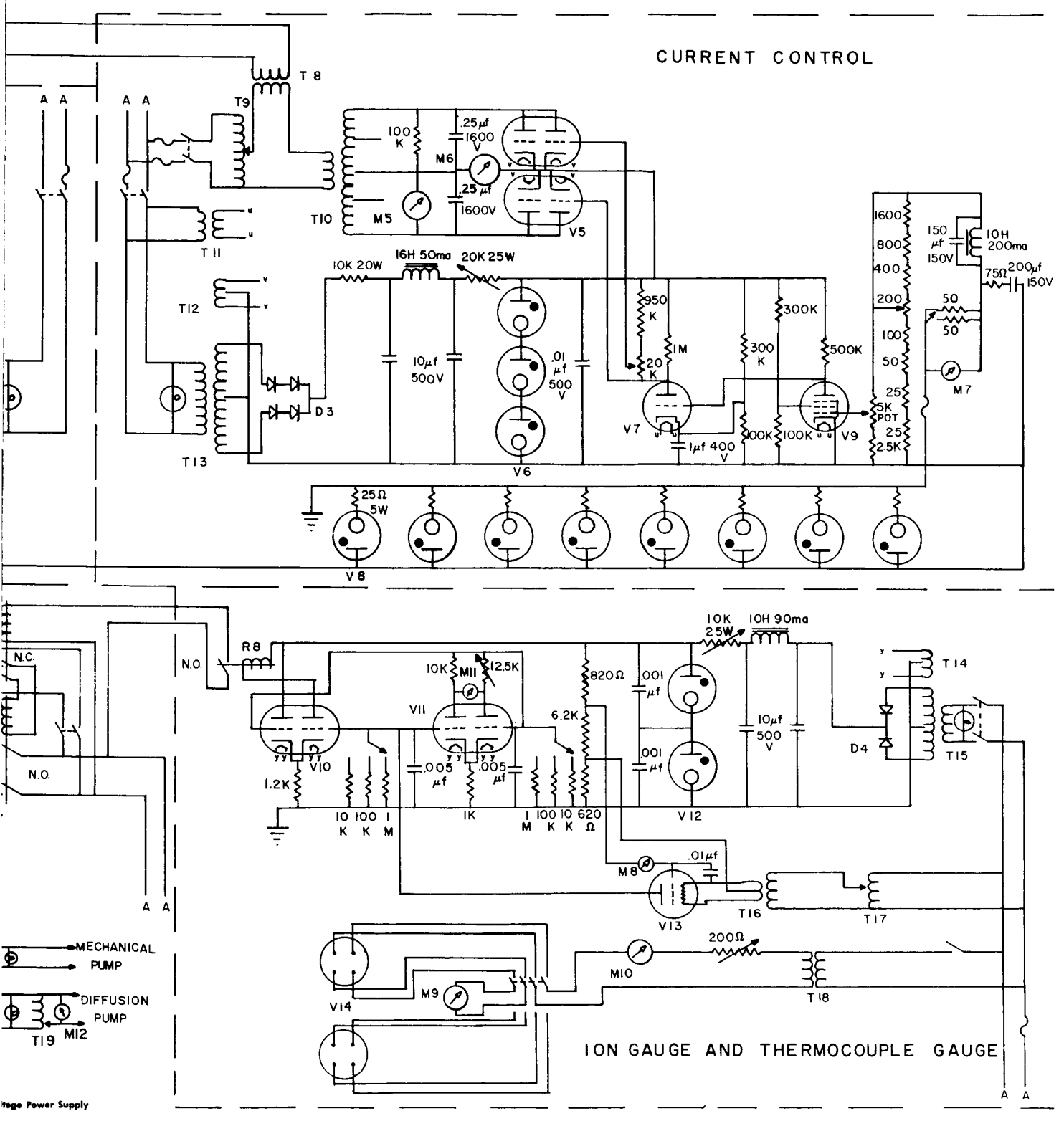
The 10 cm x-ray diffractometer described in this report has been completely operational for about two months. After the initial alignment procedures and "debugging" some preliminary data have been obtained.



Legend for High

METERS		TUBES		DIODES
M1	0-75A A.C.	V1	450TH	D1 Syntron HB2280
M2	0-50KV (0-1MA) D.C.	V2	6AU6	D2, 3, 4 IN2071
M3	0-1 MA D.C.	V3	12AX7	
M4	0-200 μA D.C.	V4	5651	**TRANSFORME**
M5	0-750V A.C.	V5	6AS7-G	T1 4:500, 15KVA R
M6	0-100MA D.C.	V6	5651	T2 1:2, 15KVA Alli
M7	0-100MA D.C.	V7	12AX7	T3 Simpson 5:1 Me
M8	0-10MA D.C.	V8	0C3	T4 General Radio V
M9	0-20μA D.C.	V9	6AU6	T5 Stancor F-725
M10	0-100MA A.C.	V10, 11	6SN7	T6, 7 Triad R-5
M11	0-1MA D.C.	V12	0C3	T8 1:1 50KV Isolati
M12	0-150V A.C.	V13	1949 Ion Gauge	T9 General Radio V
		V14	1946 Themocouple Gauge	

HIGH VOLTAGE POWER SUPPLY



tage Power Supply

ch Cottrell
ilmers 15R2310K
rf.
W50HG3

orthern Engineering
W5L

T10 Stancor P-8042
T11 Stancor P-6456
T12, 13, 14, 15 Triad R-5
T16 Stancor P-6456
T17 General Radio Variac 200B
T18 Triad F-13X
T19 General Radio Variac W5L

RELAYS
R1 General Electric CR 105E
R2, 3, 4, 5, 6 Potter Brumfield MR11A
R7 General Electric CR 105D
R8 Potter Brumfield GB5D

PRIMARY POWER
AA Regulated 115 VAC.
BB Unregulated 115 VAC.

Figure 10

The alignment of the four slit geometry is quite critical - the four slits must have their edges parallel and equally separated with the axes of the collimating slits (1 and 2) and the analyzing slits (3 and 4) perpendicular to the axis of rotation of the analyzing slits and colinear at zero scattering angle. These alignment conditions should persist for different slit widths so that a realignment is not required for each slit width used. The above alignment conditions were obtained after a preliminary alignment using optical techniques and a final alignment using the x-ray beam. In addition, once the four slits are aligned the 10 cm scattering chamber must be properly "pointed" so that the x-ray beam uniformly illuminates the sample through the first two slits. To obtain symmetric scattering the sample must be perpendicular to either the axis of the collimating or analyzing slits and to obtain optimum scattering the sample must be centered about the axis of rotation of the analyzing slits. The 10 cm scattering chamber is also hooked up so that the air in the chamber can be evacuated or replaced with He.

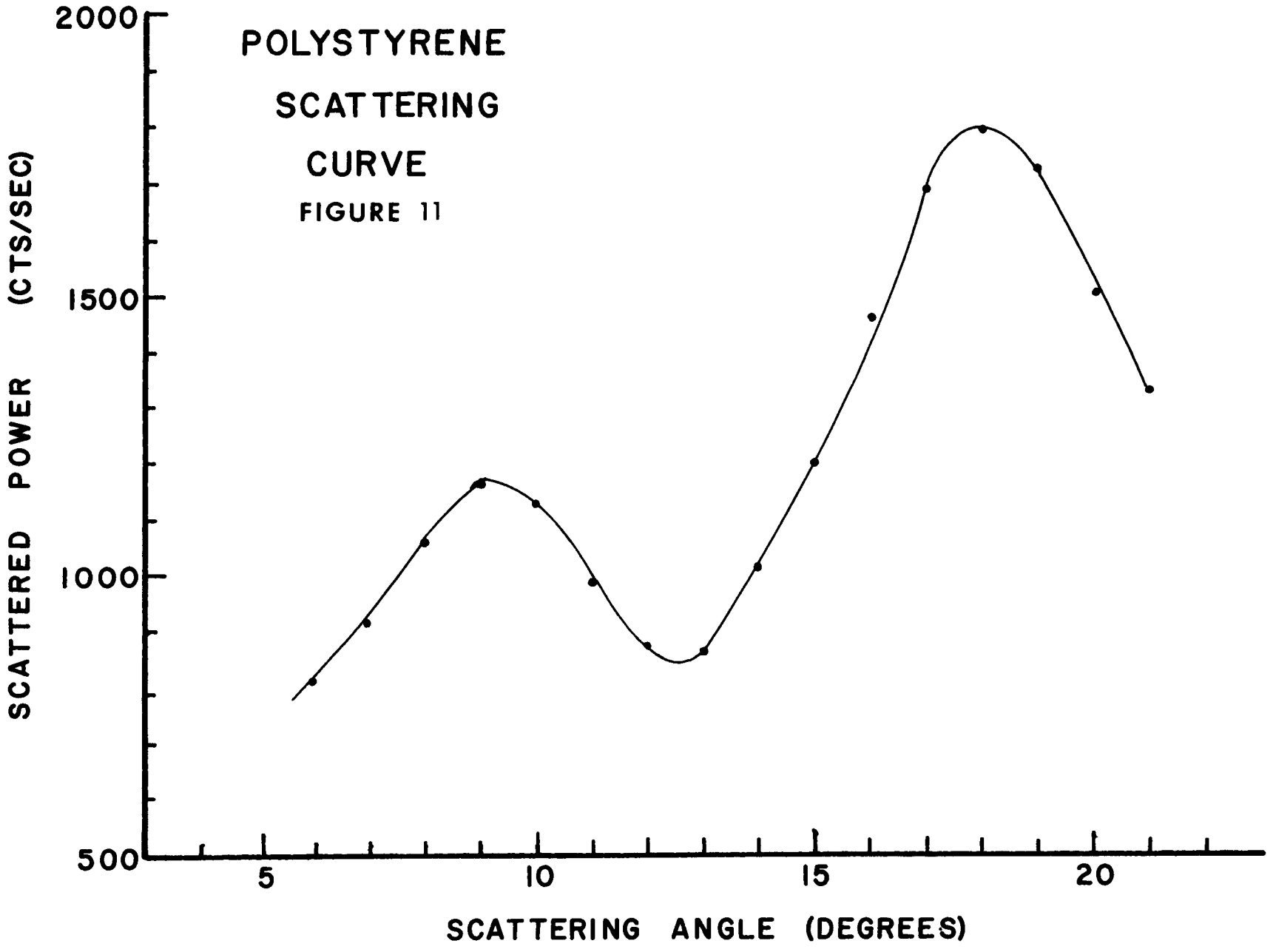
After some violent oscillations between the two 450TH series regulator tubes, now corrected with two 100 Ω resistors on the grids, the high voltage power supply is operating normally. So far it has only been loaded to 190 MA at 30 KV with most frequent operation at 30 KV, 140 MA. The rotating anode x-ray tube is also operating normally and has accumulated over 120 hours of operation.

The preliminary data obtained to date have been a series of scattering curves from plastic sheets in a search for a secondary standard scatterer for a day to day reference standard. Some sheets

of polystyrene have been found suitable for this purpose. These sheets will be calibrated against the primary standard, the gas scattering, and then used to monitor the x-ray beam intensity intermittently during a data run. A separate piece will be kept and used very infrequently as a check on radiation damage or deterioration. The polystyrene sheet is also used to measure sample transmissions. By using the 9^{o} peak as a source of K_{α} radiation the counts are accumulated for fixed time periods with the sample, whose transmission is to be measured, alternately put in and out of the scattered beam between the 4th slit and the detector tube. Figure 11 shows the scattering curve of the polystyrene sheet.

Some preliminary data have also been obtained from SF_6 gas but the sample holder used had mica windows and considerable Fe K_{α} fluorescence was observed. This problem is now under study.

Currently several important experimental problems are under investigation as described in this report and thus recommendations on the research tasks will be deferred until more data are available. One of the more serious problems to date in addition to the equipment delivery delays has been that of local power failures. The system is well interlocked so that hopefully no major damage will result from a power failure but every power failure results in the bad vacuum circuit shutting off the diffusion pump, as it should. Thus a delay occurs until the x-ray tube is again pumped down to the 10^{-5} Torr range. Six such power failures have occurred since March. The local authorities are taking steps to correct this situation.


POLYSTYRENE
SCATTERING
CURVE
FIGURE 11
2000
1500
1000
500
SCATTERED POWER (CTS/SEC)
5
10
15
20
SCATTERING ANGLE (DEGREES)

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